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Two-wave mixing in photorefractive $BaTiO_3$:Rh at 1.06 μm in the nanosecond regime

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We present two-beam coupling experiments in the nanosecond regime at 1.06 μm, using photorefractive $BaTiO_3$:Rh. The maximum observed exponential gain coefficient is 14.2 cm^{-1}. No intensity-dependent electron–hole competition and no strong saturation of the photoionized charge carriers are observed for intensities of less than 20 MW cm^{-2}. The energy required for recording the photorefractive grating is not significantly different in the nanosecond and the cw regimes. © 1997 Optical Society of America

In the cw regime, rhodium-doped barium titanate ($BaTiO_3$:Rh) exhibits a photorefractive gain as great as 23 cm^{-1} at 1.06 μm,[1] and the electrical conduction is attributed mostly to holes.[2] In this regime the crystal behavior can be analyzed, and its parameters deduced, with a three-charge-state model.[3] It has been shown that in the visible range,[4–6] at high intensities, two-beam coupling gain in undoped $BaTiO_3$ is strongly affected by an increase of pulse intensity. A saturation of the density of photoinduced charge carriers is observed, and a progressive change in the sign of the gain is explained by an intensity-dependent electron–hole competition.

We present here the results of two-wave mixing experiments in the nanosecond regime, at 1.06 μm in photorefractive $BaTiO_3$:Rh, with the goal of analyzing the effects of the pulse energy on the gain and the photorefractive time constant and determining whether the sign of the gain remains constant in this material, an experimental fact that would of course be essential for practical applications.

We first measured the variation of the two-beam coupling exponential gain coefficient Γ with illumination for a crystal referred to as Y32 B cut at 45° and with dimensions of 4.3 mm × 3.8 mm × 2.1 mm along the (100), (011), and (01$\bar{1}$) axes, respectively. This crystal was grown from a melt containing 2000 parts in 10^6 (ppm) of rhodium. The laser source was a Q-switched Nd:YAG laser that delivered 10-ns pulses at 1.06 μm with a repetition rate of 10 Hz. The coherence length is ~1.5 cm. We used the counterpropagating geometry shown in Fig. 1 in the attenuation configuration. The large grating wave number k_g provides a smaller time constant τ, which is useful for slow, weakly absorbing materials such as $BaTiO_3$:Rh at 1.06 μm ($\alpha \approx 0.15$ cm^{-1}). The attenuation configuration diminishes the oscillation of spurious beams in the crystal, which has been shown, in the cw regime, to compete with the desired photorefractive grating.[1] The experiments were conducted with extraordinary polarizations for energies ranging from 10 to 200 mJ/pulse incident upon the sample, corresponding to average illuminations per pulse of 1–20 MW cm^{-2}. The experimental data for Γ versus illuminations are plotted in Fig. 2(a). Γ is reasonably constant through the whole experimental range. No evidence is found for an intensity-dependent electron–

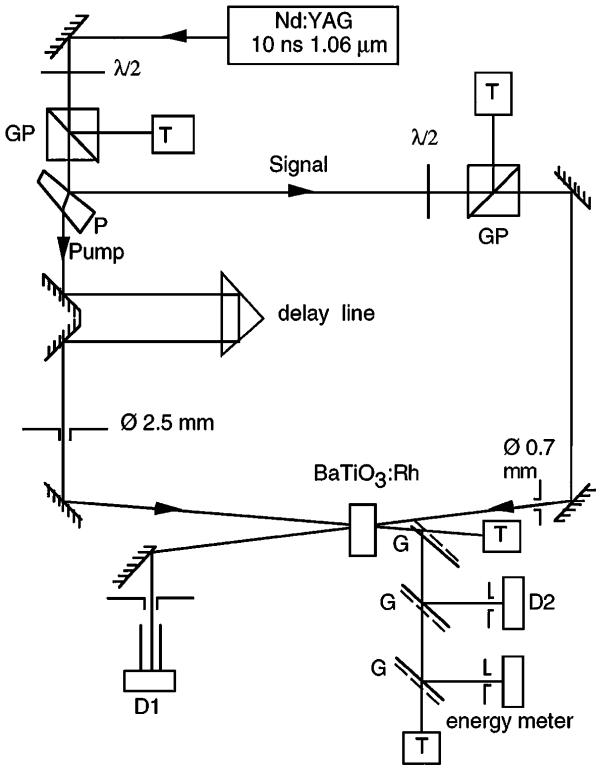

Fig. 1. Experimental setup in the counterpropagating geometry: GP's, Glan polarizers; G's, glass plates; T's, light traps; D1, D2, detectors; P, prismatic glass plate.

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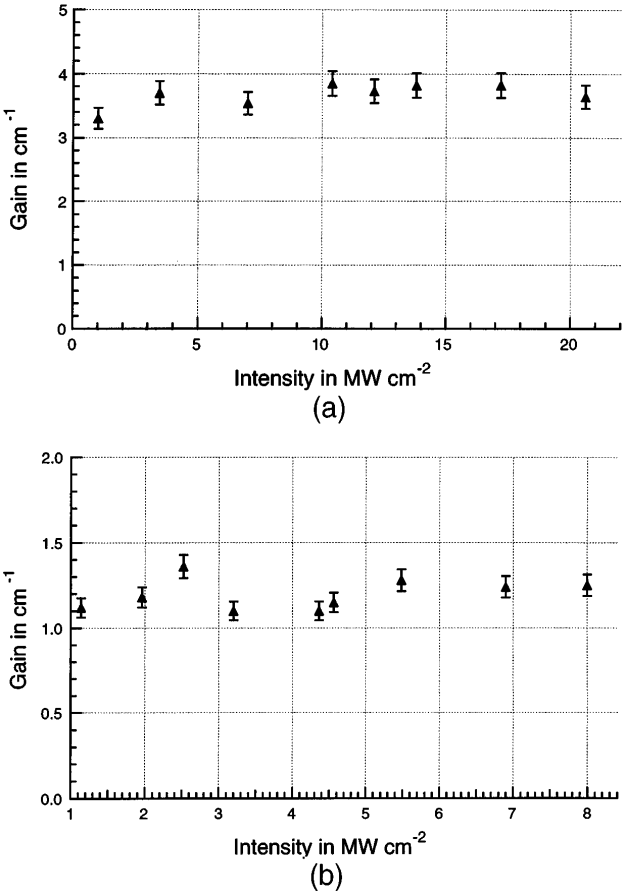

Fig. 2. Two-beam coupling exponential gain coefficient measured in the counterpropagating geometry versus intensity (a) with extraordinary polarized beams in 45°-cut Y32 B crystal and (b) with ordinary polarized beams in 0°-cut X14 crystal.

hole competition at 1.06 μm in the nanosecond regime in this wide range of intensities. This could be explained by the simple fact that a photon at 1.06 μm does not carry enough energy to excite an electron from a rhodium site to the conduction band. For a new $BaTiO_3$:Rh crystal referred to as X14 (a 4.3 mm $\times$ 3.8 mm $\times$ 2.1 mm sample cut at 0° and grown from a melt with 1000 ppm of rhodium), we also measured a nearly constant exponential gain for illuminations per pulse of 1–8 MW cm^{-2} with ordinary polarizations [Fig. 2(b)]. Therefore the single-carrier model seems suitable for describing the photorefractive effect in the nanosecond regime in $BaTiO_3$:Rh at 1.06 μm.

We observed the rise time of the photorefractive effect by recording its kinetics for different pulsed illuminations I in the counterpropagating geometry. Considering the cw-equivalent illumination I_{eq}, which has the same average energy as our nanosecond pulses at a repetition rate of 10 Hz (20 MW cm^{-2} pulsed stands for 2 W cm^{-2} in the cw regime), we deduced the cw-equivalent time constant τ_{eq} from a simplified model, which supposes that the modulation of the interference pattern is constant along the propagation axis (this is reasonably valid for small gains). The kinetics of the photorefractive gain can then be approximated by an exponential. The attenuation of the signal beam is described by

$$I_S(t) = I_S(0)\exp \Gamma l[1 - \exp(-t/\tau)],$$

where l is the interaction length and Γ is the negative exponential gain coefficient. The evolution of the time constant τ_{eq} with I is shown in Fig. 3 for the Y32 B crystal. The function $1/\tau_{eq}(I)$ seems relatively linear up to 20 MW cm^{-2}. Furthermore, the saturation parameter $f_h = S_h I \tau_h$, which defines the relative number of photoionized photorefractive traps,[5] remains smaller than 5% for $I < 20$ MW cm^{-2} when S_h, the photoexcitation cross section for holes, and τ_h, the hole recombination time, are given typical values.[3] Under such conditions, our experimental results is in agreement with Fig. 3 of Ref. 5: saturation is negligible. This might be caused by a decreased value of S_h at 1.06 μm, as suggested by the very low absorption at that wavelength. We also compared the energies that were required for the steady-state photorefractive effect in the cw and the nanosecond regimes. For both samples the product $I_{eq}\tau_{eq}$ (100 and 116 J cm^{-2} for the Y32 B and the X14 samples, respectively) is equal within the error bars for both regimes. This result does not disagree with the predictions of Ref. 4, in which it is stated that $I_{eq}\tau_{eq}$ in the nanosecond regime should be equal to or larger than $I\tau$ obtained in the cw regime.

In Y32 B crystal, we obtained the higher beam-coupling exponential gain coefficient in a copropagating configuration, with extraordinary polarized beams for an angle of 45° between the c axis and k_g, for $k_g \approx 4$ μm^{-1} (specific to this crystal).[1] The setup is identical to the one shown in Fig. 1, except that two additional mirrors are placed before the sample for an angle of 40° between the signal and the pump beams in air. We obtained an exponential gain coefficient of 14.2 cm^{-1}, uncorrected from the erasing effect caused by the reflection of the pump beam onto the rear face of the crystal, which is incoherent with the input waves. Without this effect, we would obtain $\Gamma = 16.6$ cm^{-1}. These values are smaller than those obtained in the cw regime with the same crystal.[1] In

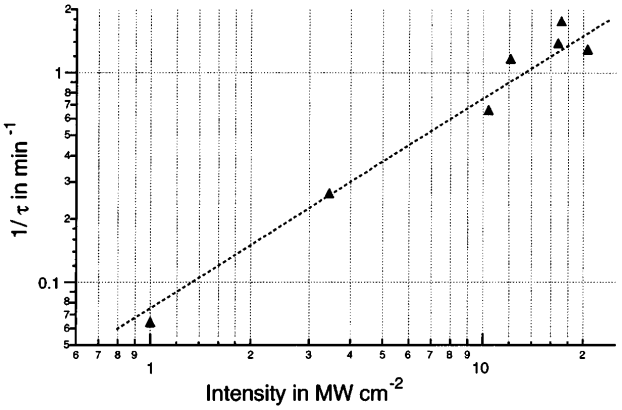

Fig. 3. Inverse of the cw equivalent time constant (proportional to the photoionized density of holes) versus pulsed intensity for Y32 B crystal. The dotted line is a guide for the eye.

our experimental conditions, i.e., with a saturation parameter $f_h < 0.05$ and a grating period $\Lambda_g \approx 1.3\Lambda_o$, where Λ_o is the optimum grating period at low intensity, Fig. 5 of Ref. 5 predicts a smaller reduction of Γ than the one we measured. We attribute most of the extra reduction of Γ to experimental problems: the control of the overlap of the beams inside the crystal imposed a large-diameter pump beam (2.5 mm), which favors the oscillation of spurious beams as observed in the cw regime.[1]

To summarize, we have demonstrated high ($\Gamma = 14.2\ \mathrm{cm}^{-1}$) two-wave mixing gain at 1.06 μm in the nanosecond regime in photorefractive $BaTiO_3$:Rh. We observed that the required averaged density of energy was roughly the same as in the cw regime. The samples do not show any intensity-dependent electron–hole competition in the range of intensities that we used (to 20 MW cm^{-2}), and the saturation of the excitation of charge carriers (holes) seems negligible. All these results are quite favorable for high-intensity applications such as correction of nanosecond pulsed laser beams at 1.06 μm.

We acknowledge the help of and numerous productive discussions with Gilles Pauliat.

Note added in proof. A paper by Brignon *et al.*[7] appeared during the process of review that reports self-pumped phase conjugation in $BaTiO_3$:Rh of a nanosecond pulsed laser beam at 1.06 μm but does not address the material characterizations presented here.

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